Study unveils gapless ground state in an archetypal quantum kagome

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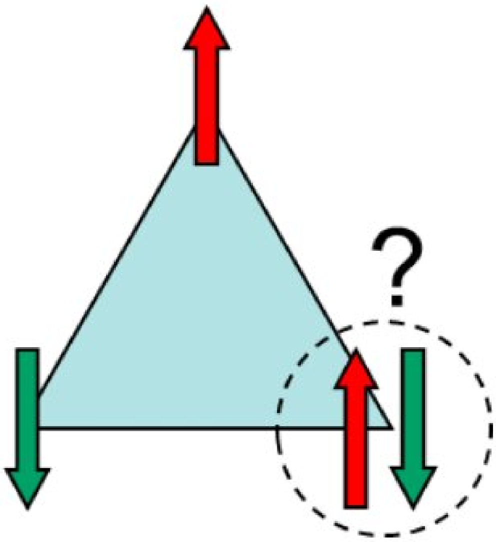


Credit: SQM group, Laboratoire de physique des solides, Orsay, Univ. Paris-Saclay.

At low enough temperatures, magnetic systems typically become solid crystals. A renowned phenomenon through which this happens is ferromagnetism, occurring when all elementary moments or spins interact at the atomic scale (i.e., the so-called Heisenberg interaction) and align in one direction. Ferromagnetism underpins the functioning of several everyday objects, including compasses, fridge magnets and hard drives.

In some cases, neighboring moments and spins can anti-align in order to minimize the pair interaction energy. When a lattice has a triangular geometry, however, this pairwise minimization becomes impossible, giving rise to a phenomenon known as 'frustration." Frustration appears to be a unique tool to defeat the paradigms of classical magnetism and let more exotic quantum states emerge.

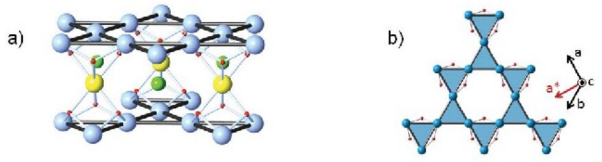
Physicists have been conducting studies aimed at determining the ground state of frustrated quantum magnets for several decades now, as this could have important implications for condensed matter physics. Building on these previous studies, researchers at Paris-Saclay University and other institutions in France have recently conducted an experiment aimed at unveiling the ground state of archetypal quantum kagome $ZnCu_3(OH)_6Cl_2$.

"On a triangular lattice, spins would classically order at an angle of 120 degrees, the best compromise in that frustrating context," Philippe Mendels, one of the researchers who carried out the study, told Phys.org. "In the 1970's Phil Anderson proposed an alternative to this best compromise when quantum effects become important, such as with half-spins, the so-called resonating valence bond state. Neighboring spins would still assemble (marry) into pairs and disassemble (divorce) to create pairs between new partners, leading to a typically fluctuating pair assembly."

The persistently fluctuating ground state theorized by Anderson is known as 'spin liquid' state, as it resembles the state observed in liquids. This is a highly entangled state with billions of spins, where individual spins lose their identity and merge into a macroscopic collective state.

"The spin liquid state idea was revived by Anderson himself as a seed to high-temperature superconductivity discovered in the 1980s," Mendels explained. "In the '90s, people started to question under which conditions this RVB state might be stabilized in antiferromagnets. Researchers soon discovered that the kagome, a David star-shaped lattice comprised of corner-sharing triangles, may be the ideal structure in which to look for spin liquids, particularly using quantum spins 1/2, which are most prone to fluctuations."

Over the past few decades, many studies focused on two simple research questions: whether stabilizing a spin liquid state on a kagome lattice is actually possible, and if so, what the most stable ground state attainable is. Evidence now suggests that achieving a spin liquid state in kagome lattices is possible, yet what the most stable state achievable is remains unclear.



Herbertsmithite structure. Credit: Khuntia et al.

"While on the experimental side, kagome materials are scarce, one of them, and still likely the best example to date, $ZnCu_3(OH)_6Cl_2$, was first synthesized in the mid 2000s and produced in a crystalline form only in the 2010s," Mendels said. "This fantastic material enables the quantum magnetism community to challenge theoretical predictions, and now pushes up our current understanding of the problem."

In their study, which was featured in *Nature Physics*, Mendels and his colleagues investigated the magnetic properties of the kagome $ZnCu_3(OH)_6Cl_2$ ground state. Their ultimate goal was to discover what class of spin liquids this material belongs to.

"Nature is not perfect, and although likely the best prototype for the kagome antiferromagnet, $ZnCu_3(OH)_6Cl_2$ still suffers from defects," Mendels said. "Zn and Cu are too similar to stay where they should ideally to produce a perfect spin-½ kagome antiferromagnet. Some Cu^{2+} spins indeed locate out of the kagome lattice and obscure the investigations, calling for standard experiments such as magnetization-specific heat."

In their experiments, Mendels and his colleagues used nuclear magnetic resonance (NMR), a technique that enables the collection of local observations and which is the basis of magnetic resonance imaging (MRI), one of the most widely used methods to detect medical conditions. Via low-temperature NMR, they were able to distinguish between defective and non-defective areas in the material in order to isolate the unique signatures of kagome spins. This procedure ultimately allowed the researchers to single out specific characteristics and dynamics in $ZnCu_3(OH)_6Cl_2$.

When trying to discriminate between different classes of spin liquids, scientists must first try to understand how pairs of spins break in a way that fits with the picture delineated by Anderson in his theories. This means determining whether there is a gap between ground and excited states, which can be more challenging when dealing with a superposition of quantum states. The study carried out by Mendels and his colleagues could be one of the first steps in this direction.

"By studying the local susceptibility, the response to a magnetic field, and the way the excitations occur when we heat the sample from temperatures close to absolute zero, we clearly show that there is no gap in the excitations energy spectrum and discuss some consistency with recent predictive theories about the excitations," Mendels said. "Whichever the final conclusion will be, we provide strong constraints to theories and narrow the range of possible models."

In their recent work, Mendels and his colleagues gathered valuable new insight about the states and characteristics of kagome materials. Overall, their findings suggest that archetypal quantum kagome $ZnCu_3(OH)_6Cl_2$ does not harbor any spin gap, which is aligned with numerical calculations conducted by other research teams. In the future, this important observation could serve as the basis for other condensed matter physics studies, ultimately broadening the current understanding of frustrated quantum magnets.

"One of our long-term dreams is to produce a highly frustrated, if not kagome, quantum material that could be doped to become a metal, meeting

Anderson's views of a novel kind of superconductor," Mendels said. "The scope of this work is even broader, as topology in condensed matter has become very popular after the 2016 Nobel prize award. Kagome-based metals are greatly sought after for their topological properties. Our work can open up new avenues of research into novel concepts, but it may also help to tackle new challenges in fundamental physics and materials science."

More information: P. Khuntia et al. Gapless ground state in the archetypal quantum kagome antiferromagnet ZnCu3(OH)6Cl2, *Nature Physics* (2020). DOI: 10.1038/s41567-020-0792-1

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